A CLASSIFICATION OF CHEMICALLY SENSITIVE SEMICONDUCTOR DEVICES

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Abstract

A general scheme is presented for classifying chemically sensitive semiconductor devices (CSSDs) CSSDs reported in the literature up to now, as well as related physicochemical phenomena, are briefly discussed and shown to fit in the scheme

1. Introduction

The impact of semiconductors on our present society would not have been realized without a thorough study of the (electro)chemical, physical, electronic and technological aspects of semiconductor materials and semiconductor devices

This paper deals with Chemically Sensitive Semiconductor Devices (CSSDs), in which all four elements mentioned above are involved, the chemistry being the most relevant As a contribution to the rapidly developing CSSD field, an attempt is made here to classify them, for this purpose a general scheme is suggested emphazising analogies (i) between related fields of semiconductor research and (ii) between CSSDs and their chemically *in*sensitive electronic equivalents

The scheme will be presented below Next, CSSDs already reported in the literature are briefly discussed and it is shown how these fit in the proposed scheme, finally, the same is done for CSSD-related physicochemical phenomena

2 Classification scheme for CSSDs

The scheme is in fact a classification of XYS-systems, in which S stands for semiconductor, X is either a metal (electronic conductor), an electrolyte (ionic conductor) or a gas, and Y is related to the properties of an insulating layer on the semiconductor For Y three cases are distinguished

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(i) The insulating layer, if sufficiently thick and of good quality, will be an insulator in the traditional sense of the word In the following a layer of this type will be denoted as I

(ii) When the thickness of the insulating layer is less than about 5 nm, it is involved in electronic conduction processes associated with tunnelling phenomena Such a layer will be denoted as i

(iii) The insulating layer may be absent

As a result the scheme incorporates autonomous and widely differing fields in semiconductor science, each of which has been intensively studied, but not necessarily from the viewpoint of chemical sensing, in this way the scheme also accounts for the versatility in applications and fundamental semiconductor research

Now it is not difficult to fill in the scheme with devices or device structures (Figs 1 and 2) Analogies should immediately be clear, of which three examples will be given

(i) MIS devices, realized either as MIS-capacitors or as MISFETs are characterized *inter alia* by their flat-band voltage V_{FB} or their threshold voltage V_T Devices with chemically active gate metals may have a threshold voltage that is dependent on environmental parameters and can thus be used for chemical sensing purposes, nevertheless, the electronic operation

	METAL (M)	ELECTROLYTE (E)	GAS (G)
THICK INSULATING LAYER (I)	MIS - structure MIS capacitor MIS field - effect transistor	EIS structure EIS capacitor EIS field effect transistor	GIS structure
THIN TUNNELABLE LAYER (i)	MiS structure MiS tunnel diode	EiS structure	GiS structure
NO INTERMEDIATE LAYER	MS structure Schottky diode	ES structure semiconductor / electrolyte junction	GS structure semiconductor / gas interface

Fig 1 Scheme for the classification of Chemically Sensitive Semiconductor Devices (CSSDs) A CSSD is to be characterized as an XYS-structure in which S stands for semiconductor, X is a metal, an electrolyte or a gas, and Y is related to the properties of an intermediate layer The elements of the matrix thus obtained are easily recognized as often well-studied fields in semiconductor research

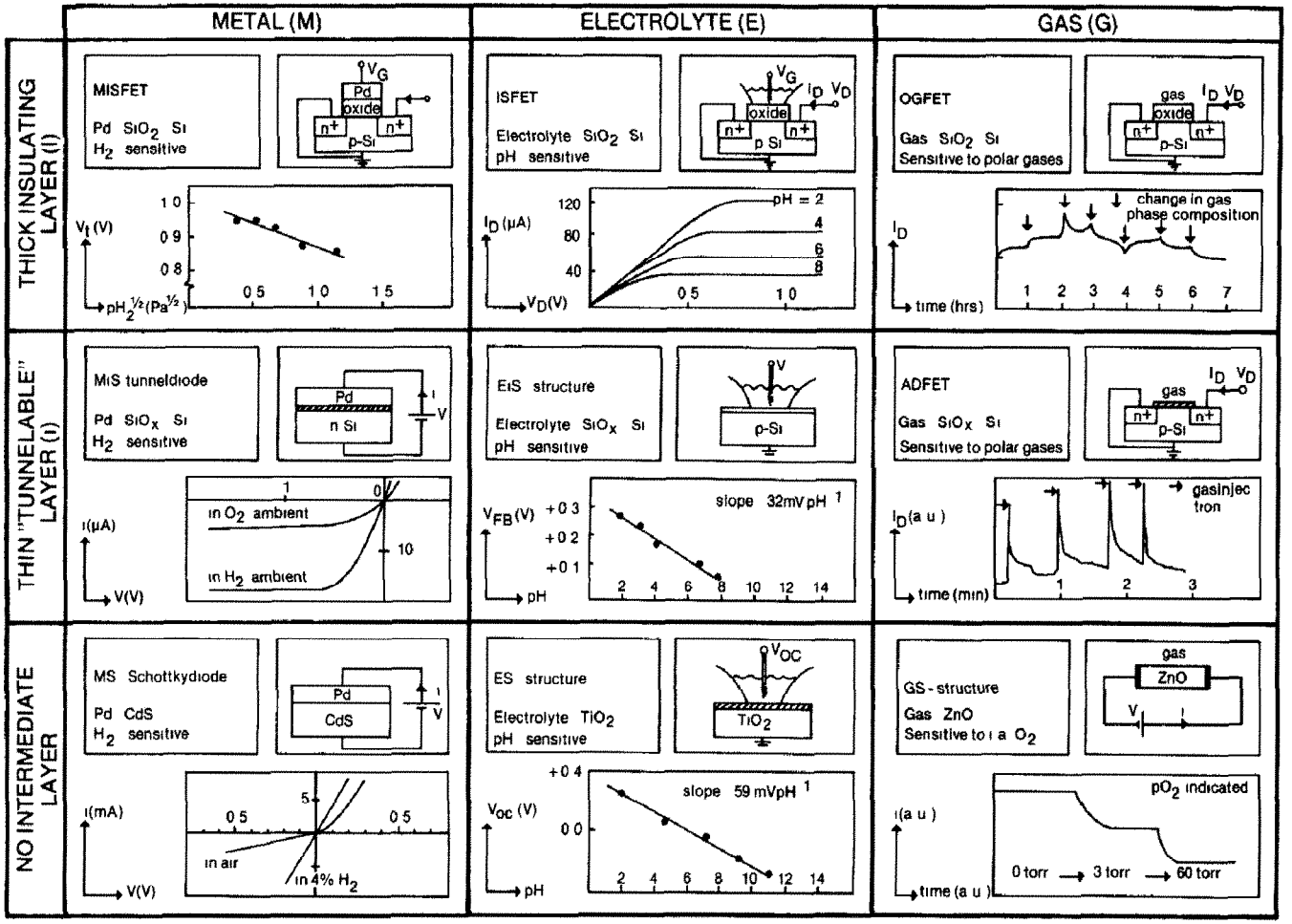

Fig 2 Classification of Chemically Sensitive Semiconductor Devices The scheme of Fig 1 is now filled with chemical sensor devices and/or principles For each matrix element, a device structure is depicted and its electric output (a characteristic current or voltage) as a function of a chemical input (an ionic concentration in a solution or a partial gas pressure) is shown schematically Data are taken from the literature These and other devices are briefly discussed in the text, where references to the original literature can be also found

principles of these chemically sensitive devices are just the same as for the chemically *in*sensitive MIS devices

EIS devices, mostly realized as ISFET structures, are also properly described by a chemically dependent threshold voltage, thus representing a clear example of the analogy between chemically sensitive MIS and EIS devices

(ii) Another striking resemblance is demonstrated by the formulae describing the current–voltage characteristics of Schottky diodes (MS structures according to Fig 1) and semiconductor/electrolyte structures (ES structures according to Fig 1) The former is given by [1]

$$I = AT^2 \exp(-\psi_B/kT)[\exp(qV/kT) - 1] \tag{1}$$

whereas the latter appears in the literature as [2, 3]

$$I_{\phi_E = 0} = AT^2 \exp(-q\phi_B/kT) \tag{2a}$$

or

$$I = I_0[\exp(q(\phi_1 - \phi_1{}^0)/kT) - 1] \tag{2b}$$

In these formulae I = current density, A = Richardson's constant, T = absolute temperature, ψ_B, ϕ_B = Schottky barrier energy and potential respectively, k = Boltzmann's constant, q = electronic charge, V = voltage across the diode, $I_{\phi_E = 0}$ = current density at zero electrode potential and $(\phi_1 - \phi_1{}^0)$ is the equivalent of V in eqn (1)

The resemblance as expressed in the above formulae is a reflection of the well-known rectifying behaviour of both structures

(iii) The analogy between solid/electrolyte and solid/gas interfaces is still poorly clarified, although the first steps in this very interesting field have already been taken an entire volume of *Surface Science* [4] is devoted to the so called 'non-traditional techniques in electrochemistry', in which information is obtained by studying the same solid surface with both (wet) electrochemical and (dry) solid state physics techniques So far, these investigations support the idea of an analogy between the columns headed 'Electrolyte' and 'Gas' in Figs 1 and 2

3. Classification of sensor devices

The elements of the matrix in Fig 1 will now be discussed from the viewpoint of CSSDs

(i) MIS–CSSDs

The 'trick' to be performed here is to replace the gate metal normally used (Al) by one that is known to possess chemical reactivity, such as the group VIII elements Ni, Pd and Pt In this way devices sensitive to hydrogen [5], carbon monoxide [6], hydrogen disulphide [7] and ammonia [8] were

realized The principle underlying the sensor operation of these devices is the formation of a dipole layer at the metal–insulator interface, the resulting voltage drop is measured as a flat-band voltage shift in either MIS capacitance–voltage characteristics or MISFETs and may equivalently be described by an environmental dependence of the metal work function

The devices may be realized with single or multilayered dielectrics [9] In MIS capacitors with Hg as the metal, large flat-band voltage shifts were observed upon the addition of solid Na to the Hg [10] Of course, this flat-band voltage shift is due to the formation of sodium amalgum, whose work function differs considerably from that of the pure mercury

(ii) MIS–CSSDs

Examples of these are the Pd–SiO_x–Si hydrogen sensor of Zemel *et al* [11] and the Al–SiO_x–Si moisture sensor of Duszak *et al* [12], where the insulator layers are no more than a few nanometres thick The former device is operated as a MIS-tunnel diode, whereas the latter is realized as a transistor-like structure, employing two interacting MIS-tunnel diodes

The fundamentals underlying both sensor structures are as yet not fully elucidated For the hydrogen sensor a chemically sensitive metal work function, thus modifying the MIS current–voltage characteristics, is suggested, extra current paths, involving tunnelling from surface states to the metal, are suggested by Zemel *et al* [11], but denied by others [13] No intensive studies on the mechanism of the above moisture sensor have been reported, but some kind of protonic involvement in the conduction processes in the silicon oxide layer is probable

It should be noted that the role of the insulator is quite different in both cases in the moisture sensor the oxide layer is likely to be involved in the chemical part of the sensing process In the hydrogen sensor, the oxide layer also plays a chemical role, but not in a sensing sense the main reason for its presence is to prevent the undesired formation of intermediate species, such as Pd_2Si, while preserving the hydrogen sensitivity of the diode structure

A MIS-hydrogen sensor has also been realized with amorphous hydrogenated silicon [14]

(iii) MS–CSSDs

Finally, with no insulating layer left between the metal and the semiconductor, we arrive at CSSDs based upon the chemical sensitivity of Schottky diodes In view of the foregoing discussion, it is not surprising to find that they consist of a catalytically active metal, almost exclusively Pd, and a semiconductor that must meet the following requirements (i) no undesired formation of intermediate species through reaction with the metal and (ii) no Fermi level pinning

Hydrogen sensors based upon Pd–ZnO [15], Pd–CdS [16] and Pd–TiO_2 [17] Schottky diodes have been reported It is commonly believed that their operation is properly described by the chemical sensitivity of the metal

work function, thus modifying the Schottky diode current–voltage characteristics

Recently, a hydrogen-sensitive photodiode was also reported [18]

MIS-, MiS- and MS-based gas sensors are extensively discussed by Lundstrom [19]

(iv) EIS–CSSDs

An intensively studied class of CSSDs is formed by EIS devices, ISFETs [20] and Ion Controlled Diodes (ICDs) [21] fall into this category Both are field effect devices and employ the electrochemical properties of the insulator–electrolyte interface, basically its response to changes in the solution pH It now seems satisfactorily established [22, 23] that these devices operate through the so called site-dissociation model, in which the acid–base equilibria at the insulator–electrolyte interface are accounted for

Using standard double-layer theory, a relation between the solution pH and ψ_0, the potential at the insulator–electrolyte interface with respect to the bulk of the solution, was derived and experimentally verified [22, 23]

With respect to the insulator, the operation of these devices is greatly improved (*i e* , nearer to Nernstian pH response, better selectivity, less drift) by using Si_3N_4, Al_2O_3 or Ta_2O_5 as an extra dielectric layer on top of the originally employed SiO_2 layer [24]

(v) CSSDs based upon EiS and ES structures

The (electro)chemical properties of semiconductor–electrolyte structures and of structures with an ultrathin insulator between the two are seldom used for chemical sensing purposes The main interest in these systems is for solar energy conversion and fundamental research in materials science and on electron transfer mechanisms Besides, applications of these systems suffer from their low stability due to electrochemical and corrosion processes at the semiconductor–electrolyte interface A vast literature [25] exists on the subject Nevertheless, observations on elemental and compound semiconductors have been made, that, at least in principle, indicate their possibilities for sensor applications

(a) The germanium/aqueous electrolyte interface was the first semiconductor/electrolyte system to be studied, starting with the work of Brattain and Garrett [26] Concerning the chemical sensor aspects, the pH of an aqueous solution could be measured provided hydrogen peroxide, H_2O_2, was present [27], a response of 60 mV pH^{-1} was found Furthermore, an interaction was found between Ge electrodes and cupric [28] and iodide [29] ions However, these observations have not led to the development of useful chemical sensors

(b) The determination of the pH response of Si electrodes is usually influenced by the formation of a very thin silicon oxide film, this explains why Madou *et al* [30] found a non-Nernstian pH response of about 30 mV pH^{-1} in the flat-band voltage determination This pH response is also found with SiO_2-gated ISFETs, where the Si surface is intentionally oxidized A

carefully designed experimental procedure and an extrapolation method enabled Chazalviel [31] to obtain a Nernstian response for the rest potential V_r in the current–voltage characteristics of a n-Si–aqueous electrolyte interface

Wolkenberg [2] performed experiments on Si–electrolyte interfaces with aqueous Na^+ and K^+ solutions, but the results are not encouraging from a sensor viewpoint A further point to be noticed is that ISFETs with only a 3 nm layer of gate oxide still behave as ISFETs, *i e* , normal transistor characteristics and a 30 mV decade^{-1} pH response are readily obtained over a surprisingly long period of time [32] This demonstrates that the use of the capacitive properties of EIS or ES systems is more promising than the use of their current–voltage behaviour

(c) With compound semiconductor electrodes the situation is not much different from that for elemental semiconductors For the semiconducting metal oxides such as ZnO, H^+ and OH^- are the potential determining ions (p d i), in principle allowing their use in pH determinations Recently, the feasibility of Pt, Ir and Ti oxides for pH determinations was discussed by Kinoshita and Madou [33] For CdS H^+ and HS^- were shown to be the p d i [34] One important application is that of the pH sensitivity of the Ir/Ir_xO_y electrode [35], which is used in such sophisticated devices as pacemakers [36]

It is interesting to notice that already in the earlier literature on this subject, the concept of a chemically sensitive semiconductor *device* is present often the semiconductor electrode is not simply a substitute for a noble metal electrode, but some kind of bipolar transistor structure is used In these so-called 'thin slice arrangements', the electrolyte replaces *e g* , the emitter of a bipolar transistor (the base and the collector are formed inside the semiconductor electrode) Ultimately, as was done by Pleskov [37], both the emitter and the collector were replaced by electrolytes, a thin semiconductor electrode thus forming the base of a transistor in which the base/emitter and the base/collector junctions were constituted by semiconductor/electrolyte junctions However, a major step forward came with the introduction of another type of transistor, namely the ISFET Speaking in terms of the scheme in Fig 1, this improvement was accomplished by going from ES and EIS structures to EIS structures

(vi) Semiconductors and oxides in contact with gases

From the beginning of semiconductor research the influence of gaseous ambients on the semiconductor surface properties has been a subject of investigation Earlier examples of these are studies of the reversible water adsorption and desorption on etched germanium surfaces by Brattain and Bardeen (the Brattain–Bardeen cycle, [38]) and of the influence of humidity on the behaviour of pn-junctions in silicon covered with a thick oxide layer by Shockley *et al* [39] In retrospect, it is surprising that the development of CSSDs was not taken up at that time

In this case too it is meaningful to classify the sensor structures according to the nature of the insulating film on the semiconductor

By omitting the gate electrode of a MOSFET, Johannessen created a gas sensing device called the OGFET (Open Gate Field Effect Transistor), the physics and chemistry of this device were more intensively studied by Thorstensen [40] The device shows sensitivity to a number of gases such as the vapours of water, alcohols, benzene and carbon tetrachloride

In Charge Flow Transistors (CFTs), developed and applied for chemical sensing by Senturia [41], part of the gate metal of a MISFET is replaced by a poorly conducting film, which may interact with its gaseous environment and thus be applied as a chemical sensor Also, its sensitivity to ions in an electrolyte could be investigated, then, the device is no longer operated in its GIS, but in its EIS mode

A gas sensing device called the Adsorption Field Effect Transistor (ADFET) was patented by Cox [42] in 1974 It is a FET-like structure with its extremely thin (<5 nm) gate oxide directly exposed to a gaseous environment, its fast and reversible response to a diversity of polar gases may be enhanced by suitable chemical surface modifications The response disappeared for devices with 20 nm-thick gate oxides It is likely that simple adsorption/desorption reactions are the main operation principle of this device

The Surface Accessible Field Effect Transistor (SAFET, [43]) is very much like the ADFET Whereas in the ADFET the gate metal is completely omitted and the device thus operates with a floating gate potential, in the SAFET a polysilicon electrode is placed a short distance above the extremely thin oxide film, thus properly defining the electric field So far only the response to water, acetone and alcohols has been shown experimentally, but extensions as with the ADFET should be possible

It is obvious that the ADFET and the SAFET are GIS-CSSDs

Compound semiconductors are becoming increasingly popular as gas sensors Generally they are brought directly into contact with a gaseous environment without the interference of a thin insulating layer as with silicon, and they thus belong to the class of GS-CSSDs Usually the conductance of a homogeneous semiconducting layer is measured as a function of environmental parameters with two electrodes, SnO_2 and ZnO in particular are intensively studied Homogeneous semiconducting gas sensors have recently been reviewed by Heiland [44] and Morrison [45]

4. Results in related physicochemical experiments

The generality of the scheme presented here also allows other physicochemical phenomena to be classified in XYS-systems Some relevant examples will be presented

(i) EIS systems, first example

In their studies of the Si/SiO_2 electrode, Madou *et al* [46] describe the electrical conducting properties of SiO_2 layers that have previously been 'activated' by a thermal or electrochemical treatment resulting in electron transfer reactions at the SiO_2/electrolyte interface, they suggest the use of these structures as 'a new class of FET-based sensors sensitive to redox couples'

(ii) EIS systems, second example

Wolkenberg [47] studied EIS systems, but with the purpose of investigating the properties of the Si/SiO_2 interface Although MIS-devices are more often used in that kind of experiments, no fundamental objections can be raised against an electrochemical approach, as is in accordance with our scheme

(iii) EiS systems

In the same paper [47] Wolkenberg calculated and experimentally determined the current–voltage characteristics of EiS systems with a 2 0 - 2 2 nm oxide layer on silicon, the results turn out to be quite analogous to those for MiS systems

(iv) GiS systems

Interesting physicochemical phenomena involving silicon–thin oxide–gas structures are reported by Caplan *et al* [48] When irradiated with u v light in the presence of oxygen, the so called P_b-centres ($Si_3 \equiv Si\cdot$ groups at the Si/SiO_2 interface) are depopulated, as monitored by a decrease in their ESR signal Optically-induced electron transfer from the P_b-centres to adsorbed oxygen molecules, thereby forming O_2^--molecules and ESR-invisible $Si_3 \equiv Si^+$ groups, is suggested as an explanation for this phenomenon In the reverse effect electron tunneling through the thin oxide layer may be involved These phenomena occur exclusively for thin, *i e* , less than 5 nm, oxide layers

5. Concluding remarks

Of course, CSSDs may be divided into groups or classes in many different ways, *e g* , according to the principle of their electronic operation (capacitive, resistive, are the devices transistors, diodes or simply resistors?) or the principle of their chemical operation (which species is actually involved in the physicochemical part of the sensing? is the sensing reaction reversible? which surfaces or interfaces play a role?)

We feel that in the scheme presented here, many of the above considerations are accommodated in a reasonable fashion In addition, the scheme is very general, allowing related fields of investigations to be included as well

The devices discussed in this paper are called chemically sensitive semiconductor devices, but in most cases the semiconductor is merely involved in

the electronic part of the sensing process, *i e* , its ability to form modifiable surface- or space-charge layers is used Only in the case of the semiconductor–electrolyte (ES) and semiconductor–gas (GS) systems is the semiconductor itself involved in the physicochemical part of the sensing process

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